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Angular Distribution of Fragments from Fission of U^{238} and Th^{232} by 45-, 80-, and 155-Mev Protons*

J. W. MEADOWS†
Cyclotron Laboratory, Harvard University, Cambridge, Massachusetts

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Measurements have been made of the angular distributions of $Sr^{91,92}$, $Ag^{112,113}$, Ba^{139} and the total fission products from proton-induced fission of U^{238} at 45, 80, and 155 Mev and Th^{232} at 45 and 155 Mev. The anisotropy decreases with increasing energy and is negative for asymmetric fission at 155 Mev.

INTRODUCTION

IN recent years the angular distributions of fragments from the fission of heavy elements have been studied by several experimenters.[1–9] Brolley and co-workers[1–3] have measured the distributions of fragments from the neutron induced fission of several heavy elements from the fission threshold to 20 Mev. They find that except near the threshold the distributions are characterized by a positive anisotropy, where the anisotropy is defined as

$$\left[\frac{\sigma(0°)}{\sigma(90°)}\right]_{\text{c.m.}}-1, \qquad (1)$$

the angle being measured with respect to the incident direction. Studies of the distributions of specific fragments from fission induced by 22-Mev protons in thorium and several uranium isotopes[4,5] also show a positive anisotropy which increases with increasing mass asymmetry. Similar results are obtained for fission induced by 42-Mev alpha particles[6] but with much larger anisotropies. Studies of photofission[7] show that near the threshold the anisotropy is negative for the total fission fragments and approaches zero with increasing energy. Furthermore, measurements of the distributions of specific fragments have shown that the anisotropy is near zero for symmetric fission but becomes increasingly negative with increasing mass asymmetry.

Recently Bohr[10] has developed a view of anisotropic fission which seems to account qualitatively for much of the above results. The preferred direction for fission should be along the nuclear symmetry axis. The orientation of this axis is dependent on the quantum numbers I, M, and K, where I is the spin of the compound nucleus, M is the projection of I on the z axis (the incident direction), and K is the projection of I on the nuclear symmetry axis. At energies several Mev. above the threshold for fission, the nucleus may pass through many different states at the saddle point; however, a marked preference for small values of K is expected. The spin of the compound nucleus is the resultant of the spin of the target nucleus, the spin of the particle, and the angular momentum of the particle. At energies of several Mev or more, the angular momentum of the particle will dominate and the preferred orientation of I will be nearly normal to the incident direction. Thus, the preferred direction for fission induced by fast particles should be parallel to the incident direction, giving a positive anisotropy.

Any process which increases or decreases the degree of orientation of I should increase or decrease the anisotropy in a corresponding fashion. Increasing the energy of the bombarding particle causes an increase in the orientation of I, although this may be largely lost due to increased neutron emission before fission. Bombarding with alpha particles should be particularly effective since the alpha particle will bring in almost twice the angular momentum as a proton of the same energy. At still higher bombarding energies, direct-interaction processes should become increasingly important and the degree of orientation of I should become steadily less. The anisotropy would be expected to pass through a maximum and eventually decline to near zero. Recent measurements of the angular distribution of specific fragments from the fission of Bi^{209} by 450-Mev protons[8] show a positive anisotropy which is much less than that observed with 42-Mev alpha particles.[6] On the other hand, Lozhkin and co-workers[9] have shown that the gross fission fragments from the fission of uranium by 660-Mev protons have a large negative anisotropy. On the basis of the above discus-

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† Now at Argonne National Laboratory, Lemont, Illinois.

[1] W. C. Dickinson and J. E. Brolley, Jr., Phys. Rev. **90**,388 (1953).

[2] J. E. Brolley, Jr., and W. C. Dickinson, Phys. Rev. **94**, 640 (1954).

[3] R. L. Henkel and J. E. Brolley, Jr., Phys. Rev. **103**, 1292 (1956).

[4] Cohen, Jones, McCormick, and Ferrell, Phys. Rev. **94**, 625 (1954).

[5] Cohen, Ferrell-Bryan, Coombe, and Hullings, Phys. Rev. **98**, 685 (1955).

[6] Coffin, Fairhall, Halpern, and Hickenlooper, University of Washington, Annual Progress Report, Cyclotron Research, 1956, 1957 (unpublished).

[7] E. J. Winhold and I. Halpern, Phys. Rev. **103**, 990 (1956).

[8] R. L. Wolke and J. R. Gutmann, Phys. Rev. **107**, 850 (1957).

[9] Lozhkin, Perfilov, and Shamov, J. Exptl. Theoret. Phys. U.S.S.R. **29**, 292 (1955) [translation: Soviet Phys. JETP **2**, 116 (1956)].

[10] A. Bohr, *Proceedings of the International Conference on the Peaceful Uses of Atomic Energy* (United Nations, New York, 1956), Vol. 2, p. 151; Suppl. Nuovo cimento **4**, 1091 (1956).

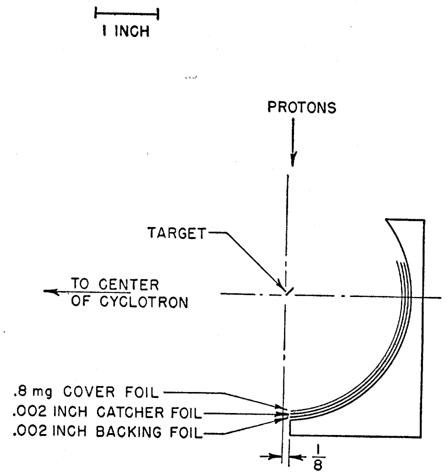

FIG. 1. Target assembly.

sion this implies a well-oriented nuclear spin at the time of fission. If the nucleus still has the expected preference for small values of K, the preferred orientation of I must be parallel to the incident direction.

In the experiment reported in the present paper an attempt is made to investigate the behavior of the anisotropy at energies above the expected maximum, to see if the anisotropy changes sign in the energy range available and to see if the dependence on mass asymmetry persists at these energies. Measurements have been made of the angular distributions of $Sr^{91,92}$ $Ag^{112,113}$, Ba^{139}, and the total fission products from proton-induced fission of U^{238} at 45, 80, and 155 Mev and Th^{232} at 45 and 155 Mev.

EXPERIMENTAL PROCEDURE

The method used was similar to that described by Cohen.[4,11] The target assembly is shown schematically in Fig. 1. The source of fission fragments was a 0.001-inch aluminum foil coated on one side with 0.5 mg/cm^2 of U_3O_8 or ThO_2 located at an angle of 45° to the proton beam. Platinum foils were used as source backings for some of the 45-Mev bombardments but could not be used at the higher energies because of platinum fission. The effective area of the source was about $\frac{1}{32}\times\frac{1}{4}$ inch. The fission fragments were caught on foil strips held 2.00 inches from the source. These were $\frac{7}{8}$ inch high and divided into segments of about 12° angular width. Seven points were measured in the forward direction, covering mean angles from 19.5° to 91°. By rotating the assembly through 180°, eight points could be measured in the backward direction, covering angles from 91° to 171°. Any smaller angles in the forward direction were prohibited by the curvature of the fragments in the cyclotron magnetic field.

When collecting total fission fragments, the catcher foils were made from 0.002-inch "Mylar" polyester foil sections cut approximately to the correct size. Small corrections in their area were determined by weighing prior to bombardment. This was necessary since the foil sections close to the beam lost up to five percent of their weight during bombardment. When the distribution of specific fragments were to be measured, the catcher consisted of a 0.002-inch aluminum foil strip which was cut into segments after bombardment. The exact areas of the segments were then determined by weighing.

In order to make certain that the activity observed was due to fragments ejected from the source during the fission process, differential range measurements were made on barium and strontium fragments at 20° and 170°. It was found that as much as 6% of the total activity was stopped in the first 0.5-mg/cm^2 aluminum absorber while only about 1.8% was stopped in the second 0.5-mg/cm^2 absorber. There was no detectable dependence on angle for either absorber. Experiments involving the electrical heating of a freshly bombarded target indicated that most of the short-range fragments could be due to the thermal evaporation of fragments stopped in the target.

These very-short-range fragments were removed by covering the aluminum catcher foils with a 0.8-mg/cm^2 aluminum foil and the plastic catcher foils with 0.00025-inch "Mylar" polyester film which had a surface density of 0.8–0.9 mg/cm^2. In the latter case the cover foil served a more important purpose of stopping nuclei knocked out of the target backing by the bombarding particles. The Na^{24} activity collected on an uncovered foil section near 0° amounted to 5–10% of the total fission activity and was very angle-dependent. In addition, large amounts of Na^{24} were knocked out of the aluminum block. This was removed by placing a 0.002-inch "Mylar" polyester foil between the block and the catcher foils. In the cases where specific fragments were detected the Na^{24} and other activities produced in the aluminum were removed by the chemical procedures. Blank runs indicated that extraneous activities were not more than 0.1% of the fission activity.

After bombardment the plastic catcher foils were set aside for about 8 hours to permit the C^{11} formed by the $C^{12}(p,pn)C^{11}$ reaction from scattered protons to decay. The foils were then counted by end-window Geiger counters. The two-counter method described by Cohen[4] was used to eliminate decay corrections. Each sample was counted twice for at least 20 000 counts. In a few cases the activities were followed for a period of twenty-four hours. There was no detectable variation of the angular distribution with time.

To determine the distribution of specific fragments, the aluminum catcher foil was removed from the holder, and the sections were cut out and weighed and then processed chemically. The barium and strontium determinations were made simultaneously. Separate

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[11] B. L. Cohen and R. B. Neidigh, Rev. Sci. Instr. **25**, 255 (1954).

TABLE I. Summary of results.

Proton energy (Mev)	Target	Fragment	Mass ratio	γ	$P_{\text{c.m.}}\times10^{14}$ (g cm/sec)	b/a	c'/a'
45	U^{238}	$Sr^{91,92}$	1.52	0.032 ± 0.006	1.83 ± 0.35		0.180 ± 0.019
		$Ag^{112,113}$	1.05	0.038 ± 0.007	1.77 ± 0.33		0.158 ± 0.026
		Ba^{139}	1.52	0.039 ± 0.004	1.47 ± 0.17		0.150 ± 0.016
		Total		0.036 ± 0.003	1.67 ± 0.14		0.173 ± 0.009
	Th^{232}	$Sr^{91,92}$	1.45	0.021 ± 0.005	1.19 ± 0.27		0.244 ± 0.017
		$Ag^{112,113}$	1.01	0.042 ± 0.006	1.91 ± 0.26		0.175 ± 0.021
		Ba^{139}	1.61	0.040 ± 0.005	1.42 ± 0.18		0.327 ± 0.018
		Total		0.045 ± 0.004	1.99 ± 0.16		0.192 ± 0.010
80	U^{238}	$Sr^{91,92}$	1.50	0.027 ± 0.005	1.55 ± 0.23	0.066 ± 0.022	
		$Ag^{112,113}$	1.04	0.030 ± 0.006	1.43 ± 0.13	0.102 ± 0.021	
		Ba^{139}	1.55	0.033 ± 0.006	1.24 ± 0.10	0.084 ± 0.024	
		Total		0.033 ± 0.003	1.51 ± 0.16	0.100 ± 0.011	
155	U^{238}	$Sr^{91,92}$	1.46	0.037 ± 0.007	2.11 ± 0.43	-0.059 ± 0.016	
		$Ag^{112,113}$	1.01	0.037 ± 0.013	1.74 ± 0.60	0.064 ± 0.028	
		Ba^{139}	1.59	0.029 ± 0.007	1.08 ± 0.26	-0.042 ± 0.013	
		Total		0.039 ± 0.004	1.79 ± 0.09	-0.036 ± 0.009	
	Th^{232}	$Sr^{91,92}$	1.40	0.043 ± 0.008	2.38 ± 0.45	-0.050 ± 0.017	
		$Ag^{112,113}$	1.04	0.050 ± 0.007	2.25 ± 0.34	0.032 ± 0.019	
		Ba^{139}	1.71	0.042 ± 0.007	1.52 ± 0.24	-0.094 ± 0.014	
		Total		0.041 ± 0.003	1.85 ± 0.13	0.012 ± 0.007	

runs were made for silver. The chemical yields were determined and the samples were counted by the above procedure.

RESULTS

At least three runs were made for each fragment at a given energy and direction. The relative activities of the individual foil sections were corrected for chemical yield and solid angle; each set was then normalized to the same total activity.

It was assumed that the angular distributions were symmetric about 90° in the coordinate system of the fissioning nucleus. Measurements at lower energies seem to confirm this.[4] This coordinate system will hereafter be referred to as the center-of-mass system although this is not strictly correct since any particles emitted before fission are not included. It was further assumed that the most probable angle between the incident direction and the motion of the center of mass was 0°. In this case, only the component of that motion parallel to the incident direction need be considered. The effect of this angle being something other than 0° has been discussed in reference 8. No such effects were seen in the experimental data.

The center-of-mass motion may be characterized by the quantity γ, where γ is the ratio of the velocity of the center of mass to the velocity of the fission fragment in the center-of-mass system. For small γ the approximate relation between γ and the angular distribution in the laboratory system is given by

$$\frac{1+2\gamma+3\gamma^2}{1-2\gamma+3\gamma^2}=\left[\frac{\sigma(0°)}{\sigma(180°)}\right]_{\text{lab}}. \tag{2}$$

An approximate value of γ was chosen, the relative activities and laboratory angles were corrected to the approximate center-of-mass values, and the corrected data were then fitted to formulas of the type

$$\sigma(\theta)=a+b\cos^2\theta, \tag{3}$$

$$\sigma(\theta)=a'+c'\cos^4\theta, \tag{4}$$

$$\sigma(\theta)=a''+b''\cos^2\theta+c''\cos^4\theta \tag{5}$$

by least-squares methods. If the anisotropy as represented by b/a, c'/a', or $(b''+c'')/a''$ was not equal in the forward and backward directions, another value of γ was chosen and the process was repeated.

The parallel component of the center-of-mass momentum can be obtained from γ by the relation

$$P_{\text{c.m.}}=P_F\gamma M/M_F, \tag{6}$$

where P_F is the momentum of the fragment in the center-of-mass system, M_F is its mass number and M is the mass number of the nucleus at the time of fission. Recent studies of the angular distributions of neutrons from fission induced by 146-Mev protons[12] indicate that about 2.5 neutrons are emitted after fission while 9 or 10 are emitted before fission. Monte Carlo calculations of the cascade process[13] indicate that at this bombarding energy the average excitation energy of the residual nucleus is about 88 Mev with 1.4 cascade particles, most probably neutrons. At a bombarding energy of 80 Mev the average excitation energy is 63 Mev with an average of 0.7 cascade particle. At a bombarding energy of 45 Mev, complete capture of the incident particle was assumed. On this basis it was estimated that the nucleus lost one neutron before fission for every 10 Mev of excitation energy, making the difference in mass

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[12] G. N. Harding and F. J. M. Farley, Proc. Phys. Soc. (London) **A69**, 862 (1956).

[13] Metropolis, Bivins, Storm, Turkevich, Miller, and Friedlander, Bull. Am. Phys. Soc. Ser. II, **2**, 63 (1957).

TABLE II. Coefficients obtained by least-squares fits of the data for the total fission fragments at 45 Mev to Eq. (5).

Target	Direction	b''/a''	c''/a''
U^{238}	0°–90°	0.032 ± 0.041	0.148 ± 0.079
	90°–180°	0.031 ± 0.039	0.140 ± 0.066
	Average	0.032 ± 0.028	0.144 ± 0.051
Th^{232}	0°–90°	0.059 ± 0.037	0.135 ± 0.056
	90°–180°	0.038 ± 0.039	0.147 ± 0.062
	Average	0.048 ± 0.027	0.141 ± 0.041

between the target nucleus and the fissioning nucleus 5, 7, and 10 for 45-, 80-, and 155-Mev bombarding energies. The fragment momentum was then computed on the basis of 170 Mev for the kinetic energy of fission.[14] The values of the parallel component of the center-of-mass momentum are given in column 6 of Table I. The errors given are obtained from the standard deviations computed from the least-squares fits. In most cases the values of the parallel momentum component at a given energy are within one standard deviation of the weighted average. The largest discrepancies are found for the total fragments from Th^{232} at 45 Mev and for Ba^{139} from U^{238} at 155 Mev which differ from the average by about 2.5 standard deviations. The ratio of the weighted average of the parallel component of the center-of-mass momentum to that of the incident proton is 1.04 ± 0.06, 0.65 ± 0.03, and 0.59 ± 0.02 for 45, 80, and 155 Mev, respectively. It appears that complete capture of the incident particle is still a good assumption for heavy nuclei at 45 Mev.

The results for the anisotropies are given in the last two columns of Table I. These are the values of b/a and c'/a' found by least-squares fits of the data to Eqs. (3) and (4). An attempt was made to fit the data to Eq. (5). In most cases the standard deviations of the coefficients were so large that no significance could be attached to the results. Only in the case of the total fission fragments at 45 Mev could reliable results be obtained. These are listed in Table II. For these two it appears that the $\cos^4\theta$ term is dominant, which is in agreement with the results from fast-neutron-induced fission.[2] For this reason all the 45-Mev data were fitted to Eq. (4). The values of the coefficients obtained by fitting the 80- and 155-Mev data to Eq. (5) gave no indication that either term was dominant. Since the anisotropy and γ (although to a lesser extent) are not very sensitive to the form of the distribution chosen, the 80- and 155-Mev data were finally fitted to Eq. (3).

The anisotropies measured at 45 Mev are in qualitative agreement with those obtained with 22-Mev protons[5] and 42-Mev alpha particles[6] insofar as the dependence on tha mass asymmetry is concerned. Thorium shows a strong dependence while uranium shows little if any. The actual values of c'/a' are much less than the corresponding values obtained with 42-Mev alpha particles while only a little greater than those obtained for 22-Mev protons. This is consistent with the assumption that the anisotropy is dependent on the degree of orientation of the spin of the compound nucleus. The maximum angular momentum expected from the capture of a 42-Mev alpha particle is almost twice that expected from capture of a 45-Mev proton while the excitation energies of the compound nuclei are about the same.

No measurements were made on Th^{232} at 80 Mev. Angular distributions of fragments from U^{238} targets have anisotropies that are definitely smaller than those observed at 45 Mev. Furthermore, there is some indication that at this energy symmetric fission is more anisotropic than asymmetric fission.

The anisotropies of the specific fragments at 155 Mev show a decided dependence on mass asymmetry. The asymmetric fragments have definite negative values of b/a. In all four cases these values are negative by three to six standard deviations. Symmetric fission, as represented by silver, has values of b/a which are positive by about two standard deviations.

The available results on the anisotropy of the total fission fragments from proton-induced fission of U^{238} are plotted in Fig. 2. It should be noted that these data are obtained from three sources which do not measure quite the same quantity. In the present work, only those fragments which have half-lives such that they give appreciable counting rates eight or nine hours after bombardment are measured. At 660-Mev, measurements were made on fission tracks in uranium-loaded emulsions.[9] Here, all fission fragments contribute to the measurements. The quantity actually determined was the distribution in projected angle. While it is not possible to obtain a unique distribution in spatial angle from a given distribution in projected angle, a form which will give the experimental results is

$$1-0.86\sin^2\theta+1.44\sin^4\theta. \qquad (7)$$

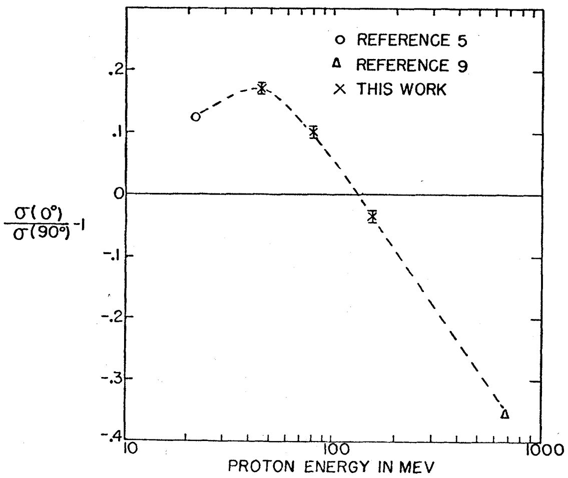

FIG. 2. Anisotropies of total fission fragments from proton-induced fission of U^{238}.

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[14] Cohen, Cohen, and Coley, Phys. Rev. **104**, 1046 (1956).

Simpler distributions as represented by Eqs. (3) or (4), adjusted to give the experimental anisotropy in projected angle, have nearly the same anisotropy in spatial angle although the resulting form of the distribution in projected angle is somewhat different. The anisotropy for the total fission products from fission induced by 22-Mev protons[5] is estimated from measurements made on five specific fragments.

The anisotropy behaves as expected at the lower energies going through a maximum in the vicinity of 45 Mev and thereafter decreasing. However it does not appear to approach any limiting value at higher energies but decreases as a smooth function of energy to quite large negative values.

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Cross Section for the $Al^{27}(\gamma,2p)Na^{25}$ Reaction to 65 Mev*

L. B. Aull and W. D. Whitehead
University of Virginia, Charlottesville, Virginia

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The $Al^{27}(\gamma,2p)Na^{25}$ cross section has been determined from 25 to 65 Mev with respect to the $Cu^{63}(\gamma,n)Cu^{62}$ cross section. The cross section has a maximum value of 0.29 mb at 32 Mev and the integrated cross section from 25 to 64 Mev is 2.8 Mev mb.

INTRODUCTION

The (γ,n) and (γ,p) cross sections have been determined for a number of elements, and the parameters describing the giant resonances as functions of A have been well systematized.[1] Several $(\gamma,2n)$ cross sections have been investigated[2,3] and the cross sections for some multiple processes have been measured,[4,5] but no $(\gamma,2p)$ reactions have been investigated in detail.[6] We have determined the cross section for the $Al^{27}(\gamma,2p)Na^{25}$ reaction, which has a threshold at 21.4 Mev, from 25 Mev to 65 Mev using the bremsstrahlung from the University of Virginia synchrotron, by measuring the induced Na^{25} activity in pure aluminum disks.[7] The yield function, determined with respect to the $Cu^{63}(\gamma,n)Cu^{62}$ cross section, was unfolded by means of the Leiss-Penfold matrices[8] to give the cross section as a function of energy.

EXPERIMENTAL PROCEDURE

To determine the aluminum yield curve, an aluminum disk and a copper disk were clamped together, the pair was irradiated for three minutes at each energy, and then the induced activity in each sample was determined. The samples were irradiated at 1-Mev intervals in the maximum photon energy range 25 to 36 Mev and at 2-Mev intervals in the range 36 to 65 Mev. The aluminum disks were 0.845 g/cm^2 thick and $1\frac{1}{2}$ inches in diameter, and the copper monitor disks were the same diameter and 0.133 g/cm^2 thick; the sample holder was 41 cm from the internal tungsten target and was centered with the aid of x-ray plate exposures. The beam was monitored with an ionization chamber also, and for each irradiation the sample was placed in the beam by remote control when the intensity was steady. If the intensity as determined with the ionization chamber fluctuated during a run, the run was discarded.

The synchrotron, which was built by the General Electric Company,[9] has a maximum energy of 65 Mev, and the gamma-ray intensity at this energy from a 0.020-inch internal tungsten target measured with a Victoreen R-meter in a $\frac{1}{8}$-inch lead thimble is 300 r/min at 1 meter from the target. The energy of the electrons was determined by means of an integrating fluxmeter,[10] and the energy scale of the machine was established with reference to breaks in the $C^{12}(\gamma,n)C^{11}$ yield function at 19.10 and 19.55 Mev.[11] The estimated uncertainty in the maximum photon energy values obtained from the integrator is ±0.25 Mev.

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[1] G. R. Bishop and R. Wilson, *The Nuclear Photoeffect*, Encyclopedia of Physics (Springer-Verlag, Berlin, 1956), Vol. 13, p. 332.

[2] A. O. Hanson and E. A. Whalen, Phys. Rev. **89**, 324 (1953).

[3] A. I. Berman and K. L. Brown, Phys. Rev. **96**, 83 (1954).

[4] Harrington, Katz, Haslam, and Johns, Phys. Rev. **81**, 660(A) (1951).

[5] Schupp, Colvin, and Martin, Phys. Rev. **107**, 1058 (1957).

[6] Davidson, Patro, and Woldseth, Bull. Am. Phys. Soc. Ser. II, **2**, 351 (1957).

[7] Supplied through the courtesy of W. C. Saunders, Reynolds Metal Company.

[8] A. S. Penfold and J. E. Leiss, Phys. Rev. **95**, 637(A) (1954).

[9] Elder, Gurewitsch, Langmuir, and Pollock, J. Appl. Phys. **18**, 810 (1947).

[10] J. E. Leiss, National Bureau of Standards (private communication).

[11] B. M. Spicer and A. S. Penfold, Phys. Rev. **100**, 1375 (1955).